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The attached documents are exact copies of the European patent application conformes à la version described on the following page, as originally filed.

Les documents fixés à cette attestation sont initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr.

Patent application No. Demande de brevet n°

99307316.2

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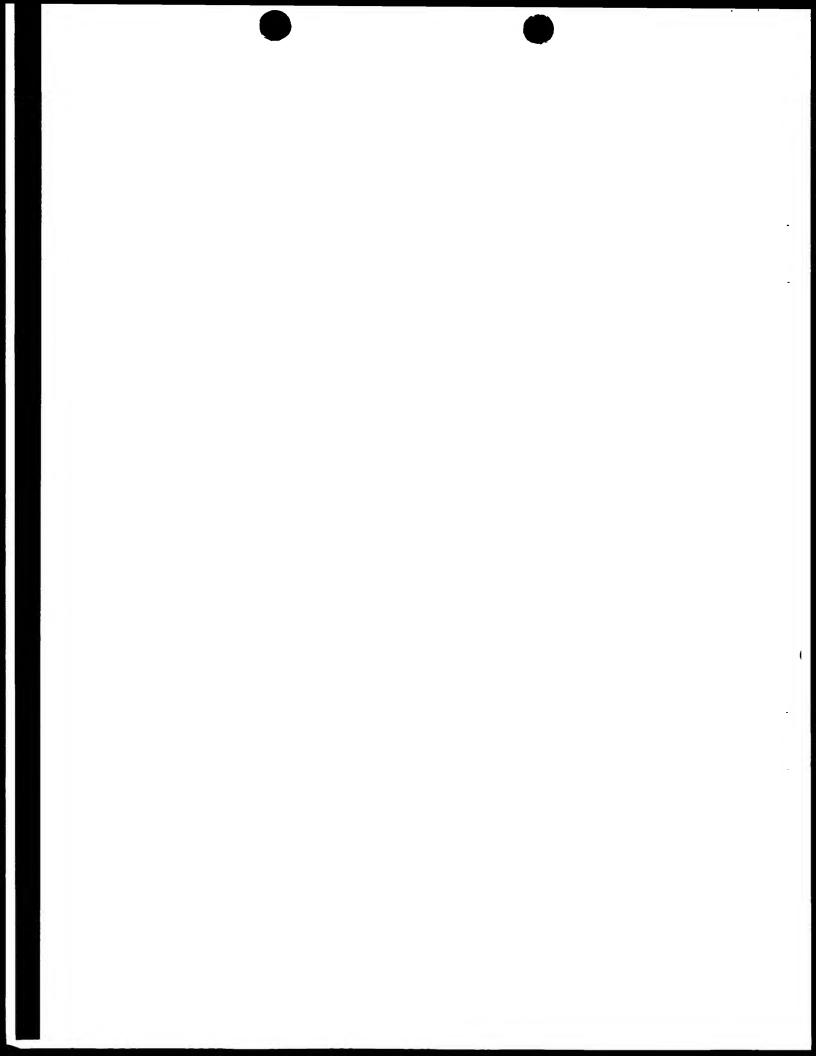
For the President of the European Patent Office

Le Président de l'Office européen des brevets p.o.

I.L.C. HATTEN-HECKMAN

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# Blatt 2 der Bescheinigung Sheet 2 of the certificate Page 2 de l'attestation

Anmeldung Nr Application no Demande n\*

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Bezeichnung der Erfindung Title of the invention Titre de l'invention

Separation apparatus

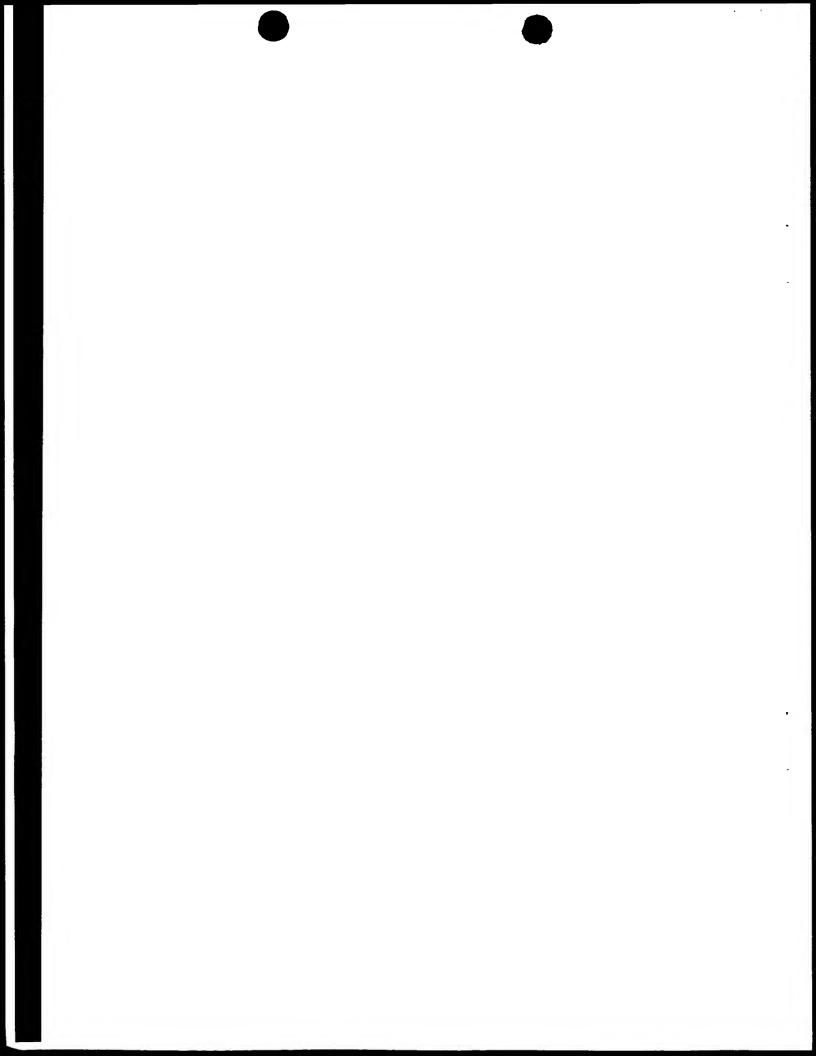
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Bemerkungen Remarks Remarques



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#### SEPARATION APPARATUS

#### Field of the invention

The invention is directed to an improved separation apparatus, wherein particles can be efficiently separated from a gas-particles mixture. The invention is also directed to the use of such an apparatus in a fluid catalytic cracking process.

#### Background of the invention

The field of fluid catalytic cracking (FCC) has undergone significant development improvements due primarily to advances in catalyst technology and product distribution obtained therefrom. With the advent of high activity catalysts and particularly crystalline zeolite cracking catalysts, new areas of operating technology have been encountered requiring even further refinements in processing techniques to take advantage of the high catalyst activity, selectivity and operating sensitivity. Of particular concern in this field has been the development of methods and systems for separating the hydrocarbon product from the catalyst particles, particularly from a high activity crystalline zeolite cracking catalysts, under more efficient separating conditions so as to reduce the overcracking of conversion products and promote the recovery of desired products of a FCC operation. US-A-4588558, US-A-5376339, EP-A-309244, US-A-5055177 and US-A-4946656 all describe developments concerned with the rapid separation and recovery of entrained catalyst particles from the hydrocarbon products. The rapid separation is achieved in that the catalyst are separated from the reactor riser effluent in a first cyclone separator of which gas outlet conduit is

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in fluid connection with a secondary cyclone. This cyclone line up in FCC reactors, sometimes referred to as close-coupled cyclone separation, minimises the residence time in which the hydrocarbon product is still in contact with the catalyst after it leaves the reactor riser.

Both in the FCC reactor, as discussed above, and in the FCC regenerator such series of primary and secondary cyclones exist. EP-A-309244 describes an example wherein coupled cyclone separators are both used in the reactor as well as the regenerator vessel. Typically more than one of these series of cyclones exist in parallel. Examples of other processes in which a primary and secondary cyclone arrangements are used are the Methyl tert-butyl ether (MTBE)-fluidized bed dehydrogenation process and in the acrylonitrile process.

There is an on-going effort to improve the separation efficiency of cyclone separation apparatuses.

One known method of improving the separation efficiency of a single cyclone apparatus is achieved by increasing the gas velocity at the inlet or at the gasoutlet of the cyclone. Although the separation efficiency of this primary cyclone shows an improvement, the overall efficiency of the first plus second cyclone separator is not improved.

Cyclone separators having a vertical tubular housing and an gas-outlet conduit having a gas-inlet opening located at about the level of an elevated cyclone roof are described Chemie Ingenieur Technik (70) 6 1 98, pages 705-708. Favourable separation efficiencies are reported for these cyclone separators.

### Summary of the invention

The object of the present invention is to provide an apparatus, wherein particles can be efficiently separated from a gas-particles mixture, which has an improved overall separation efficiency.

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This object and other objects which will become apparent when reading the description is achieved with the following apparatus.

Apparatus for separating solid particles from a suspension of solid particles and gas, wherein the apparatus comprises:

- (i) a vertical primary cyclone vessel having a tubular housing comprising of a tubular wall section provided with a tangentially arranged inlet for receiving the suspension, a dipleg at the lower end of the tubular wall section, fluidly connected to the tubular wall section by means of a frusto conical wall section, and a cover which closes the upper end of the tubular wall section, wherein the cover is provided with an axial circular opening which opening serves as a gas inlet opening of a gas outlet conduit;
- (ii) one or more secondary gas-solids cyclone separators which are fluidly connected with the gas outlet conduit of the primary cyclone.

Applicants have now found that the combined primary and secondary separation means of the apparatus of the invention achieve a far better separation efficiency that the state of the art separation devices comprising conventional primary and secondary cyclone separators. With conventional cyclone separators is here meant having a design in which the gas outlet conduit significantly protrudes the cyclone housing from above. With significantly protruding the cyclone housing from above is especially meant that the protrusion equals between 0.4 and 1.2 times the height of the tangentially arranged feed-inlet opening. A typical conventional cyclone is exemplified in Fig. 20-106 of Perry's Chemical Engineers' handbook, McGraw Hill, 5th ed., 1973.

With respect to the disclosed efficiency of the cyclone separators disclosed in the above cited article

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in Chemie Ingenieur Technik it is surprising that the combination of a primary cyclone and a secondary cyclone separation means of the claimed apparatus show such a high separation efficiency when a suspension containing relatively a high content of solids is fed to the primary cyclone. This is especially surprising in view of the fact that it is not always obvious which measures will positively influence the overall separation efficiency of a coupled cyclone separation apparatus as illustrated above for the gas inlet and outlet velocities in the primary cyclone separator. Applicants have now found that this overall efficiency can be significantly improved. In one example the particle content was reduced ten fold in the gas stream leaving the secondary separation means.

The invention is also directed to a fluidized catalytic cracking process making use of said apparatus. The invention shall be described in more detail below, including some preferred embodiments.

Detailed description of the invention

The apparatus according to the invention can find use in any process in which solid particles are to be separated from a suspension of said solid particles and a gas. Examples of such process are the afore-mentioned MTBE-fluidized bed dehydrogenation process, the acrylonitrile process and fluid catalytic cracking (FCC) process. In a FCC process solid catalyst particles can be separated from gasses in both the reactor as well as the regenerator making use of the apparatus according to the invention. On the reactor side catalyst are to be separated from the hydrocarbon product gasses. It is important that such a separation can be performed in an efficient manner making use of such a close-coupled separation apparatus. Any solids which are not separated will have to be separated further downstream, for example by making use of filters. By improving the separation

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smaller filters can be used. On the regenerator side catalyst particles will have to be separated from the flue gas leaving the regenerator. The amount of particles in the flue gas should be low for environmental reasons and to protect downstream equipment, like for example expansion turbines.

# Brief Description of the drawings

Figure 1 represents a partly cross-sectional presentation of a close-coupled cyclone apparatus according to the invention in a FCC reactor configuration.

Figure 2 represents a partly cross-sectional presentation of a preferred secondary cyclone.

Figure 3 represents a FCC reactor vessel comprising the apparatus according to the invention.

### Detailed description of the drawings

Figure 1 represent a preferred embodiment of the apparatus according to the invention. In the Figure a reactor riser (1) of a fluidized catalytic cracking process is shown which is fluidly connected via conduit (2) to a primary cyclone (3). In the Figure only one primary cyclone separator is shown for clarity reasons. Typically more than one, suitably two or three, primary cyclone separators (3) will be in fluid communication with the down stream end of a reactor riser (1). The primary cyclone (3) has a tubular housing (4) consisting of a tubular wall section (5) provided with a tangentially arranged inlet (6) for receiving the suspension of catalyst particles and hydrocarbon vapour which leave the reactor riser (1). The inlet can have for example a circular or rectangular form. The lower end of the tubular wall section (5) is fluidly connected by means of a frusto conical wall section (7) to a dipleg (8). Through dipleg (8) most of the catalyst particles will be discharged downwards. The

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upper end of the tubular wall section (5) is provided with a, suitably flat, cover (9). Cover (9) is provided with an axial circular opening (10) which opening serves as a gas inlet opening of a gas outlet conduit (11). The diameter of the gas inlet opening of the gas outlet conduit is preferably between 0.3 and 0.6 times the diameter of the tubular wall section of the cyclone housing. Essential to the present invention is that the gas outlet conduit (11) does not, or not significantly, protrudes the cyclone housing from above. In a preferred embodiment of the present invention a small protrusion is allowed. Preferably such protrusion is smaller than 0.5 times the diameter of the gas inlet opening or axial circular opening in the roof of the cyclone. The gas outlet conduit (11) is in fluid communication with a secondary gas-solids separator (12). In the Figure only one secondary separator is shown for clarity reasons. In a typical arrangement more than one, suitably two, secondary separators (12) are in fluid communication with the gas outlet conduit (11) of one primary cyclone (3). The secondary separator (12) shown in the Figure is a typical cyclone separator having a gas outlet conduit (13) which protrudes the roof (14) of the tubular cyclone housing (15). Through this gas outlet conduit the hydrocarbon vapours which are poor in catalyst particles are discharged from the apparatus according the invention. The vapours are further processed in downstream product separation equipment (not shown). The secondary cyclone (12) is further provided with a dipleq (16) to discharge separated catalyst particles downwards.

Preferably the gas inlet opening of the gas outlet conduit (11) of primary cyclone (3) is located at a distance (d2) above the centre of the tangentially arranged inlet opening (6), which is greater than any

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typical values for the state of the art cyclones having a protruding gas outlet conduit. More preferably the ratio of this distance (d2) and the diameter (d1) of the tubular housing (4) is between 0.5 and 1.5. In this preferred embodiment the gas inlet opening of the gas outlet conduit (11) is flush with the cyclone cover (9).

Figure 2 shows a preferred embodiment wherein the secondary gas-solids separator is a cyclone separator (17) having a tubular housing (18) consisting of a tubular side wall (19) provided with a tangentially arranged inlet (20) for receiving the suspension of catalyst particles and vapour. To the lower end of the tubular wall (19) a dipleg (21) is fluidly connected by means of a frusto conical wall section (22). A cover (23) closes the upper end of the tubular wall (19). The cover (23) is provided with an axial circular opening (24) which opening is the gas inlet opening of a gas outlet conduit (25). Because the gas inlet opening of the gas outlet conduit (25) is flush with cover (23) less surface area is present onto which particles can adhere onto. Particle adherence is especially a problem in FCC operations where it can initiate coke formation. Coke formation on the outside wall of the protruding gas outlet conduit is a well known problem to occur in secondary FCC cyclone separators. By eliminating this surface in this preferred embodiment less catalystadherence and thus coke formation will occur. Furthermore an even more improved separation efficiency is observed.

More preferably the gas inlet opening of the gas outlet conduit (25) of the secondary cyclone (17) of Figure 2 is located at a distance (d3) above the centre of the tangentially arranged inlet opening (20) and wherein the ratio of this distance (d3) and the diameter (d4) of the tubular housing (18) is between 0.5 and 1.5.

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Figure 3 represents a preferred application of the apparatus according to the invention. Figure 3 shows a fluidized catalytic cracking (FCC) reactor vessel (26) comprising an apparatus according to the invention (28). Preferably a downstream part of a vertical elongated reactor riser (27) is present within the vessel. Alternatively such riser part and optionally also the primary cyclone may well be placed outside the reactor vessel as is exemplified in above referred to US-A-5376339. Figure 3 further shows that the downstream end of the reactor riser (27) is in fluid communication with the tangentially arranged inlet (29) of the primary cyclone (30). The reactor vessel (26) further comprises at its lower end a stripping zone provided (30) with means (31) to supply a stripping medium to a dense fluidized bed (32) of separated catalyst particles. Stripping medium can be any inert gas, steam or steam containing gasses are suitably used as stripping medium.

The reactor vessel (26) further comprises means to discharge stripped catalyst particles from the vessel via conduit (33). Via conduit (33) stripped, or also referred to as spent catalyst, is transported to a regeneration zone (not shown). In such a regeneration zone coke is removed from the catalyst by means of (partial) combustion. Regenerated catalyst is transported to the upstream part of the reactor riser where it is contacted with a hydrocarbon feed to yield the earlier referred to suspension of catalyst particles and hydrocarbon product vapours at the down stream part of the reactor riser.

The reactor vessel (26) further comprises means to discharge the hydrocarbon and stripping medium vapours from the vessel via conduit (34). Typically the gas outlet conduit(s) (35) of the secondary separation means (36) are in fluid connection with a plenum (37) from which the hydrocarbon product vapours are discharged

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together via conduit (34). In a preferred embodiment an opening (38) is present in the gas outlet conduit connecting the primary cyclone with the secondary separation means. Through this opening stripping medium and hydrocarbons which are stripped from the catalyst can be discharged from the vessel.

Examples of FCC processes in which the apparatus according the invention can be suitably used are described in the afore mentioned patent publications and those described in Catalytic Cracking of Heavy Petroleum Fractions, Daniel DeCroocq, Institut Francais du Petrole, 1984 (ISBN 2-7108-455-7), pages 100-114. Preferably the apparatus is used in a FCC process wherein a gas solids suspension if fed to the primary cyclone having a solids content of between 1 and  $12 \text{ kg/m}^3$ .

The invention shall be illustrated by the following non-limiting examples.

#### Example 1

To a separation apparatus as described in Figure 1 a gas-solids suspension was fed having the properties as listed in Table 1. The dimensions of the primary cyclone were so chosen that d1 was 0.3.m and d2 was 0.19.m.

Table 1.

average particle size (micron)	76
density of suspension $(kg/m^3)$	5.8
primary cyclone inlet velocity (m/s)	10
separation in-efficiency after primary cyclone	0.04%
Separation in-efficiency after primary cyclone	0.046
separation in-efficiency after secondary	
cyclone	0.4 ppm
pressure-drop (Pascal)	2500

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## Comparative Experiment A

Example 1 was repeated except that the primary cyclone was one of a state of the art design having a gas outlet conduit which protrudes downwardly through the roof of the cyclone housing. The bottom of the tangentially inlet and the opening of the gas outlet conduit were in the same horizontal plane. The top of the tangentially inlet and the roof of the cyclone housing in the same horizontal plane. The distance between the centre of the tangentially inlet and the frusto conical wall section was the same as in Example 1. Furthermore the inlet velocity, the composition of the suspension and the dimensions of the secondary cyclone were the same. The results are presented in Table 2.

Table 2.

Average particle size (micron)	76
density of suspension $(kg/m^3)$	5.8
primary cyclone inlet velocity (m/s)	10
separation in-efficiency after primary cyclone	0.9%
separation in-efficiency after secondary cyclone	3 ppm
pressure-drop (Pascal)	2200

## Comparative Experiment B

Experiment B was repeated except that the primary cyclone was the same state of the art design however improved in efficiency by a re-arranging of the inlet-ducting of the first cyclone with respect to the feeding riser. All other dimensions and operational data were kept the same. The results are presented below in Table 3.

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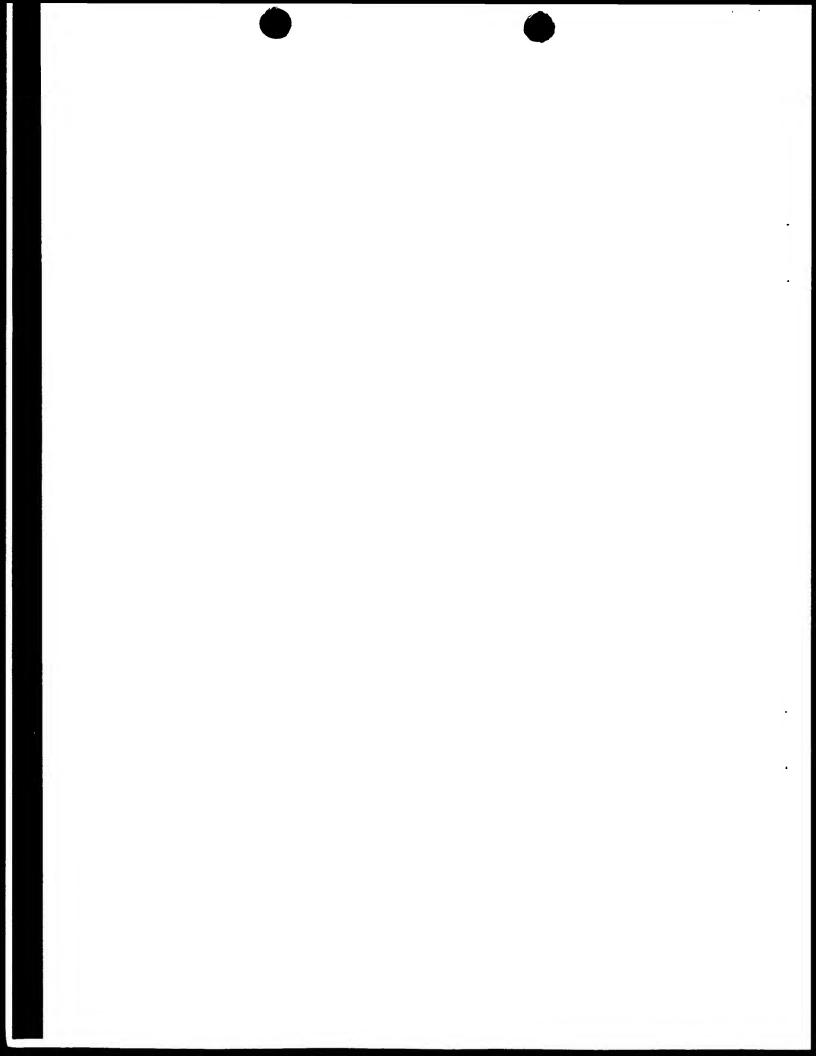
Table 3.

Average particle size (micron)	76
density of suspension (kg/m <sup>3</sup> )	5.8
primary cyclone inlet velocity (m/s)	10
separation in-efficiency after primary cyclone	0.3%
separation in-efficiency after secondary cyclone	3 ppm
pressure-drop (Pascal)	2300

By comparing the results from Example 1 with Experiment A and Experiment B with A it is apparent that in both situations the separation efficiency is enhanced after the primary cyclone. However with the cyclone having the inlet modification in combination with a secondary cyclone the total efficiency remains the same as in Experiment A. However when the apparatus according to the invention is used a significant improvement in total separation efficiency is observed when compared to the total separation efficiency of Experiment A.

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## CLAIMS

- 1. Apparatus for separating solid particles from a suspension of solid particles and gas, wherein the apparatus comprises:
- (i) a vertical primary cyclone vessel having a tubular housing comprising of a tubular wall section provided with a tangentially arranged inlet for receiving the suspension, a dipleg at the lower end of the tubular wall section, fluidly connected to the tubular wall section by means of a frusto conical wall section, and a cover which closes the upper end of the tubular wall section, wherein the cover is provided with an axial circular opening which opening serves as a gas inlet opening of a gas outlet conduit;
- (ii) one or more secondary gas-solids cyclone separators which are fluidly connected with the gas outlet conduit of the primary cyclone.
- 2. Apparatus according to claim 1, wherein the gas inlet opening of the gas outlet conduit is located at a distance (d2) above the center of the tangentially arranged inlet opening and wherein the ratio of this distance and the diameter of the tubular housing (d1) is between 0.5 and 1.5.
- 3. Apparatus according to any one of claims 1-2, wherein the secondary gas-solids separator is a cyclone separator having a tubular housing consisting of a tubular side wall section provided with a tangentially arranged inlet fluidly connected to the gas outlet conduit of the primary cyclone, a dipleg, fluidly connected to the lower end of the tubular wall section by means of a frusto conical wall section, and a cover which closes the upper end of the tubular wall section, wherein the cover is

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provided with an axial circular opening which opening is the gas inlet opening of a gas outlet conduit.

- 4. Apparatus according to claim 3, wherein the gas inlet opening of the gas outlet conduit is located at a distance (d3) above the centre of the tangentially arranged inlet opening and wherein the ratio of this distance and the diameter of the tubular housing (d4) is between 0.5 and 1.5.
- comprising an apparatus according to any one of claims 1-4, wherein the downstream end of a reactor riser is in fluid communication with the tangentially arranged inlet of the primary cyclone, the vessel further comprising at its lower end a stripping zone provided with means to supply a stripping medium to a dense fluidized bed of separated catalyst particles, means to discharge stripped catalyst particles from the vessel and means to discharge the hydrocarbon and stripping medium

Fluidized catalytic cracking reactor vessel

6. Vessel according to claim 5, wherein the gas outlet conduit of the primary cyclone is provided with an opening to receive stripping medium and stripped hydrocarbons.

vapours from the vessel.

- 7. Use of an apparatus according to claims 1-4 to separate solid particles from a suspension of particles and gas.
  - 8. Use of an apparatus according to any one of claims 1-4 or a vessel according to any one of claims 5-6 in a fluid catalytic cracking process, wherein a gas solids suspension if fed to the primary cyclone having a solids content of between 0.5 and 15 kg/m $^3$ .

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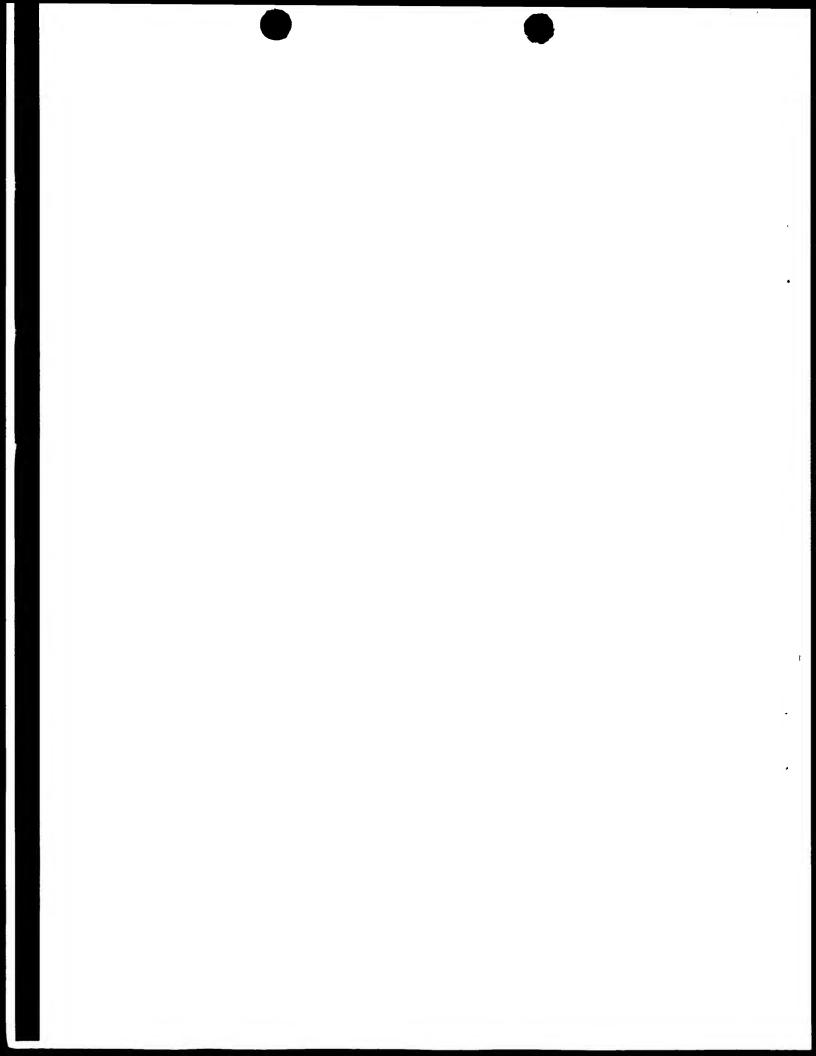
#### ABSTRACT

#### SEPARATION APPARATUS

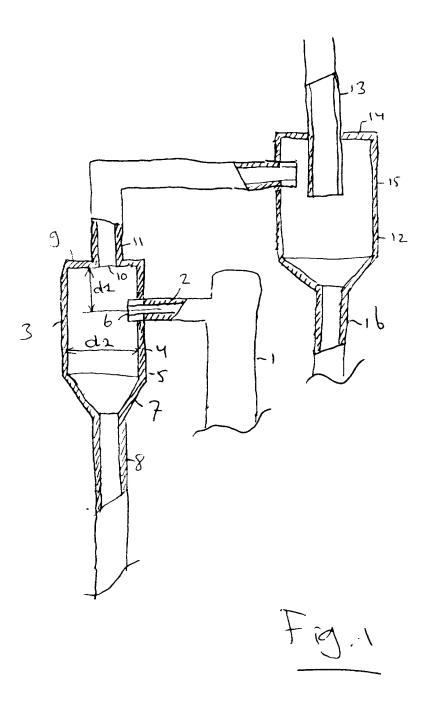
Apparatus for separating solid particles from a suspension of solid particles and gas, wherein the apparatus comprises:

- (i) a vertical primary cyclone vessel having a tubular housing comprising of a tubular wall section provided with a tangentially arranged inlet for receiving the suspension, a dipleg at the lower end of the tubular wall section, fluidly connected to the tubular wall section by means of a frusto conical wall section, and a cover which closes the upper end of the tubular wall section, wherein the cover is provided with an axial circular opening which opening serves as a gas inlet opening of a gas outlet conduit;
- (ii) one or more secondary gas-solids cyclone separators which are fluidly connected with the gas outlet conduit of the primary cyclone.

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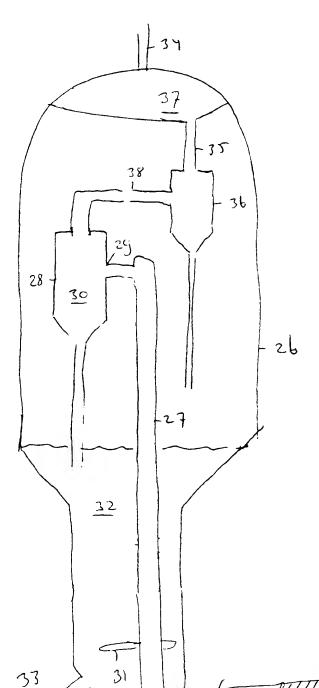


Fig.3

Fig. 2